Biological archives reveal contrasting patterns in trace element concentrations in pelagic seabird feathers over more than a century

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Abstract

Contamination of diverse environments and wild species by some contaminants is projected to continue and increase in coming decades. In the marine environment, large volumes of data to assess how concentrations have changed over time can be gathered from indicator species such as seabirds, including through sampling feathers from archival collections and museums. As apex predators, Flesh-footed Shearwaters (*Ardenna carneipes*) are subject to high concentrations of bioaccumulative and biomagnifying contaminants, and reflect the health of their local marine environment. We analysed Flesh-footed Shearwater feathers from Australia from museum specimens and live birds collected between 1900 and 2011 and assessed temporal trends in three trace elements of toxicological concern: cadmium, mercury, and lead. Concentrations of cadmium increased by 1.5% per year (95% CI: +0.6, +3.0), while mercury was unchanged through the time series (-0.3% per year; 05% CI: -2.1, +1.5), and lead decreased markedly (-2.1% per year, 95% CI: -3.2, -1.0). A reduction in birds' trophic position through the 20th century, and decreased atmospheric emissions were the likely driving factors for mercury and lead, respectively. By combining archival material from museum specimens with contemporary samples, we have been able to further elucidate the potential threats posed to these apex predators by metal contamination.

Keywords: *Ardenna carneipes*; Flesh-footed Shearwater; mercury; lead; cadmium; Western Australia

Capsule: Cadmium in Flesh-footed Shearwater feathers increased between 1900 and 2011, while mercury remained stable and lead decreased.

Introduction

Contaminants in marine and atmospheric environments are generally increasing over recent time, and are projected to increase in coming decades (AMAP, 2011; Hoffman et al., 2003; Lamborg et al., 2014; Pacyna and Pacyna, 2001; Streets et al., 2009; UNEP, 2002). With an increase in toxicological studies of a variety of organisms, and generally poor knowledge of effect thresholds in many species, understanding how contaminant concentrations have changed over time is critical to an ecological interpretation and the subsequent management and conservation efforts. If a species has high concentrations of a contaminant contemporarily, is this because it has always experienced such concentrations and has adapted to accommodate them, or have concentrations in the environment increased, and been accumulated by biota? Reporting high concentrations of a given contaminant must therefore be accompanied by appropriate interpretation, including the historical context (Bond et al., 2015).

As top predators in the marine environment, seabirds can be exposed to high levels of bioaccumulated and biomagnified pollutants from both natural, and anthropogenic sources (Burger and Gochfeld, 2002; Day et al., 2012). Seabirds can also act as indicators or sentinel species for examining the health of the marine environment, including chemical contamination (Burger and Gochfeld, 2004; Monteiro and Furness, 1995), and contaminant concentrations are monitored using seabirds in a variety of oceanic domains (Barrett et al., 1996; Braune, 2007; Burgess et al., 2013; Carravieri et al., 2016; Day et al., 2006; Goodale et al., 2008).

Feathers are used frequently to measure contaminants in seabirds (Burger, 1993). They can be sampled without sacrificing the individual, or from preserved museum skins, making studies of long time series possible. Feathers often contain the biologically active form of many metal contaminants (e.g., most mercury is in the form of methylmercury, Bond and Diamond, 2009b), and can be a significant metabolic pathway for contaminant elimination (Braune and Gaskin, 1987; Burger, 1993). Many elements in feathers, however, represent external contamination from the lithosphere, even after vigorous washing (Borghesi et al., 2016), but at present it cannot be partitioned from endogenous deposition.

A variety of studies have investigated temporal trends in contaminants, primarily mercury, using seabird feathers (Appelquist et al., 1985; Bond et al., 2015; Monteiro and Furness, 1997; Thompson et al., 1993b; Thompson et al., 1992; Vo et al., 2011). With improvements in analytical methods, reliable metal and metalloid concentrations can be acquired with about 15-25 mg of tissue (Bond and Lavers, 2011; Friel et al., 1990; Haynes et al., 2006), making archival studies more attractive, and complementary to indirect time series of seabirds' contamination through sediment cores (Sun and Xie, 2001). Museum samples are also the only way to examine time series of contamination retrospectively, which can inform contemporary conservation and management (Bond et al., 2015).

Flesh-footed Shearwaters (*Ardenna carneipes*) are trans-equatorial migrants that breed on islands in New Zealand, South and Western Australia, and on Île Saint-Paul in the Indian Ocean (Lavers, 2015; Roux, 1985; Waugh et al., 2013). Samples collected in the 2008-2009 austral summer indicated that some metal and metalloid concentrations in feathers could be of toxicological concern (Bond and Lavers, 2011), likely because of their diet of predatory fish and squid (Gould et al., 1997). Flesh-footed Shearwaters also ingest large quantities of plastic marine debris, which adults offload to their nest-bound chicks during feeding (Hutton et al., 2008; Lavers and Bond, 2016b; Lavers et al., 2014), and these plastics could provide a route for hydrophobic contaminants, and compounds used in plastic production (Holmes et al., 2012; Lavers and Bond, 2016a; Lavers et al., 2014; Tanaka et al., 2013), though the proportional contribution of many plastic-transported contaminants is unknown (Bakir et al., 2016). The question remains, however – are contaminant concentrations in Flesh-footed Shearwater feathers increasing or decreasing, and are the high concentrations reported by Bond and Lavers (2011) typical of shearwaters' contaminant burden? This is particularly germane given the significant trophic declines in shearwaters from Western Australia over the last century (Bond and Lavers, 2014), and the species' range-wide population decline (Jamieson and Waugh, 2015; Lavers, 2015; Reid et al., 2013).

Our goals were, therefore, to describe changes in trace elements in Flesh-footed Shearwater feathers over more than a century, to compare historic concentrations with those from contemporary samples, and to discuss these results in the context of contaminants in the marine environment.

Materials and Methods

Sample collection

We sampled feathers from Flesh-footed Shearwater skins housed in museum collections in Australia, Canada, France, New Zealand, and the United States (see Acknowledgements for a list of institutions). Only specimens with precise years of collection were sampled. Breast feathers were selected because they are the best indicator of whole-body metal burdens (Furness et al., 1986), and allowed comparisons with contemporary samples (most museum collections only permit sampling breast feathers). Other tissues commonly sampled from museum specimens (mainly toe pads, but also nails) are used for other purposes (e.g., genomic or isotopic analyses) and are not collected from live birds so for examination of contaminant trends over centennial scales, breast feathers are the most available and appropriate tissue (Bond et al., 2015). In addition, Flesh-footed Shearwaters replace their breast feathers during the latter half of the breeding season (February-April), before wing feathers are moulted on the wintering grounds (Onley and Scofield, 2007); given the varying temporal lags of incorporation into feathers, we cannot link measured concentrations to local exposures as we do not know the age of individual feathers within the moult cycle, and assume that feather elemental concentrations are integrated across a similar time and space among individuals. Feathers were stored in sterile polyethylene bags or paper envelopes at -20°C prior to analysis. The elements considered here are bound to the keratin protein in feathers, and do not sublimate below 60 ºC, so the possibility that some would have been lost from historic specimens during storage is remote.

As many of the museum skins were collected at sea away from breeding colonies as scientific specimens, or were taken as fisheries bycatch, their colony of origin was unknown. Using a combination of biogeochemical markers, Lavers et al. (2013) assigned samples of unknown provenance to a breeding area of origin. We used contemporary samples and archival material assigned to locations in Western and South Australia (n = 123), or known to originate there (n = 43).

Analytical Methods

Cadmium (Cd), mercury (Hg) and lead (Pb) concentrations were assessed using the same procedures as described in Bond and Lavers (2011). Feathers were washed in 0.25M NaOH to remove external contamination (Bearhop et al., 2000; Bond and Diamond, 2009a), and two feathers per bird were pooled as individual feathers can be highly variable in metal concentrations (Bond and Diamond, 2008). Trace element concentrations were measured in a PerkinElmer ELAN DRCII ICP-MS and the protocol used was based on Friel et al. (1990). Procedural blanks and secondary reference materials were included for every 15-20 samples. The secondary materials used were certified human hair samples 6H-09 and 7H-09 from the Centre de Toxicologie du Québec, Institut National de Santé Publique du Québec (Table S1). Mercury was used in museum preservation, contaminating specimens collected < 1940 with inorganic Hg (Bond et al., 2015; Vo et al., 2011). We therefore assessed temporal trends of Cd and Pb from 1900-2011, and Hg from 1946-2011 only. Recovery of the secondary reference material ranged from 89-112% among these elements for all runs (Table S1). Values were corrected for background levels using procedural blanks, and for recovery using values from secondary reference materials within each run.

Statistical Methods

We used two approaches to examine temporal changes in metal concentrations. First, we used the program PIA (version 05/11/13; Bignert, 2013) to analyse the time-series of each element, which enabled us to make comparisons with similar studies of elemental concentrations over time, most of which are from the Arctic (Bignert et al., 2004; Rigét et al., 2011). PIA uses a robust regression and log-linear regression techniques to detect linear and nonlinear trends using a running-mean smoother based on annual geometric means (Fryer and Nicholson, 1993). We performed separate analyses for each element, set the statistical power to detect a trend at 80%, and the minimum slope to detect at 10% over 10 years at $p < 0.05$ using a three year running-mean smoother.

We also applied general additive models (GAMs; Wood, 2017) in the package *mgcv* (Wood, 2019) where contaminant concentration was a function of a cubic regression spline of collection year. The number of knots was determined by generalized cross-validation in the model fitting process, and resulted in $k = 9$ for all three trace elements.

All concentrations are expressed as parts-per-million (ppm, µg/g) on a fresh weight basis. Though stable isotope data exist for this time series as well (Bond and Lavers, 2014), there is a temporal mismatch between the integration periods of $\delta^{13}C$ and $\delta^{15}N$, and trace elements in feathers so they do not reflect the same periods (Bond, 2010), and do not change the trace element concentrations measured.

Results

There was a significant linear, increase in Cd in shearwater feathers from 1900-2011 of 1.5% per year (95% CI: +0.1, +3.0%, $F_{1, 34} = 4.64$, $p = 0.037$), and ranged from 0.006-20.082 µg/g (geometric mean: 0.354 µg/g, SD: 2.924). Based on the variance among years, 29 years of data would be required to detect an annual change of 10% with 80% power. Our time series had 99% power to detect a 10% change over the entire period, and the lowest detectable annual change was 7.1% (Table 1). The GAM fit the data well ($r^2 = 0.54$), and the spline term was significant (effective df: 8.66, $F = 22.65$, $p < 0.001$), remaining relatively flat until the mid-1980s where it rose rapidly and then declined to the previous level (Figure S1).

We found no significant trend in feather Hg from 1946-2011 ($\beta = -0.3$, 95% CI: -2.1, +1.5%, $F_{1,22} = 0.12$, $p = 0.73$), though we had very high power to detect a change (94% power to detect a 10% change over the time series, and a lowest detectable change of 8.3%); 21 years of data would be required to detect an annual change of 10% with 80% power (Table 1). Feather Hg concentrations ranged from 1.290-113.499 µg/g (geometric mean: 8.241 µg/g, SD: 18.807). The GAM fit was lower than for Cd ($r^2 = 0.12$), and the cubic regression spline shoed a dip in the 1990s before rising rapidly in the early 2000s and returning to pre-1990s levels (Figure S2).

Pb in shearwater feathers decreased significantly from 1900-2011 by 2.1% per year (95% CI: -3.2, -1.0, $F_{1,24} = 27.85$, $p < 0.001$), and this time series had 100% power to detect a 10% change. An annual change of 10% could be detected with 26 years of data, and the lowest detectable change of the time series was 76% (Table 1). Overall feather Pb concentrations ranged from 0.009-1125.733 μg/g (geometric mean: 1.656 μg/g, SD: 82.411), though the upper extreme is likely influenced by external contamination. Removing this individual, concentrations ranged from 0.009-255.432 μg/g (geometric mean: 1.592, SD: 21.373). Like Hg, the GAM for Pb with only a cubic regression spline for year did not fit the data well ($r2 = 0.07$). The spline featured a peak in the late 1940s followed by a gradual decline and stabilization after 1985 (Figure S3).

Discussion

It is important to note that the age of the individuals from museum collections were unknown. Flesh-footed Shearwaters, like many in the family Procellariidae, cannot be aged based on plumage (Onley and Scofield, 2007), so our sample may include a combination of birds > 1 year old, which have undergone a complete moult, and those < 1 year old, which would still retain feathers grown at the breeding site before fledging, and therefore differ in exposure to contaminants (Braune and Gaskin, 1987; Monteiro and Furness, 2001a; b). This difference in the pool of trace elements that could be deposited into feathers (and feather age ranging from perhaps a few weeks to several months) would result in increased variance, but unfortunately cannot be controlled (Burger, 1995; Malinga et al., 2010; Stewart et al., 1999).

Flesh-footed Shearwaters in Western Australia migrate to the northern Indian Ocean (Lavers et al., 2019; Powell, 2009; Shuntov, 1968), an area with potentially concerning concentrations of toxic elements in the water column (Danielsson, 1980; Kar et al., 2008). Understanding where exposure occurs, seabirds’ roles in nutrient and contaminant transport (Blais et al., 2005; Doughty et al., 2016), and the potential carry-over effects of contaminant exposure on the non-breeding grounds (Fort et al., 2014) can inform conservation actions, and inform interpretations of population trends.

Cadmium

Little of birds' Cd burden is sequestered into feathers (Burger, 1993; Honda et al., 1985), and museum specimens can be a mechanism for monitoring the fraction depurated in feathers (Borghesi et al., 2016; Pilastro et al., 1993) so while feathers are not suitable for assessing total Cd burden, they are appropriate for examining temporal changes. A large portion of Cd in the environment comes from anthropogenic sources, including steel production and waste incineration (Hutton, 1983), and exposure is highly influenced by ocean cycling (Macdonald et al., 2005). Squid, a common prey of Flesh-footed Shearwaters, often have high concentrations of Cd (Gerpe et al., 2000; Gould et al., 1997). The most commonly identified squid in Flesh-footed Shearwaters' diet, *Ommastrephes bartramii*, had liver Cd concentrations of 287 ± 202 µg/g in the 1970s, which was higher than sympatric *Loligo opalescens*, but lower than *Symplectoteuthis oualaniensis* (Martin and Flegal, 1975), though Western Australia shearwaters' diets can also be dominated by pilchards (*Sardinops sagax*; JLL unpublished data). Cd in pilchards has not been assessed in Australia (Padula et al., 2016), but concentrations of 0.5 µg/g dry weight in muscle have been reported elsewhere (Tawfik, 2013).

Seabirds may also be able to tolerate higher concentrations of Cd (Scheuhammer, 1987). The toxicological effects of Cd on birds include kidney lesions, altered behavior, eggshell thinning, and more (Furness, 1996). These are, however, effects measured on internal organs, and so concentrations at which effects manifest range from 0.1-2.0 µg/g fresh weight (fw) in feathers (Burger, 1993; Burger and Gochfeld, 2000b). Of the 166 birds sampled here, 119 (72%) had feather Cd concentrations >0.1 µg/g, and 29 (17%) had concentrations >2.0 µg/g (Figure 1), with four individuals exceeding 10 µg/g Cd in feathers, which is among the highest recorded in wild birds (Anderson et al., 2010; Burger and Gochfeld, 2000c; Hindell et al., 1999). The peak identified in the GAM in the 1990s is interesting given that world cadmium production has remained relatively stable from 1990-2012, around 20,000 metric tons (U.S. Geological Survey, 2015).

This is the first study to examine changes in Cd in birds over time, and given the significant increase in feather Cd, and high concentrations in some recent individuals, further study on the potential sources, effects, and causes of these concentrations is warranted. The application of stable isotopes of Cd (and Hg) as tracers could be particularly beneficial in answering these questions (Conway and John, 2015; Day et al., 2012).

Mercury

Most Hg in the environment is from anthropogenic sources that are transformed into the biologically active methyl Hg, and subsequently bioaccumulated and biomagnified in food webs (Krabbenhoft and Sunderland, 2013; Lindberg et al., 2007; Weiner et al., 2003). Hg is acquired through birds' diet, and mostly eliminated in proteinaceous tissues, such as egg components or feathers, where it binds to disulfide bonds between cysteine molecules (Bond and Diamond, 2009b; Crewther et al., 1965; Monteiro and Furness, 2001a; Thompson, 1996). While contamination near point sources can be a concern for seabirds (Finger et al., 2015), global atmospheric transport and mobile predators and prey mean that Hg affects upper trophic predators, like Flesh-footed Shearwaters, regardless of location. Concentrations of Hg in feathers > 20 μg/g are thought to be of concern to piscivores (Burger and Gochfeld, 1997; Cristol et al., 2012; Evers et al., 2014), though it seems some species, notably albatrosses, are able to tolerate much higher concentrations without observed adverse effects (Bustamante et al., 2016; Hindell et al., 1999). We found 23/137 Flesh-footed Shearwaters (17%) exceeded 20 μg/g, and ranged as high as 113 μg/g in one individual sampled in 2006. Temporally, though the GAM identified a drop in the late 1990s followed by a rapid increase and levelling off of Hg concentrations in shearwater feathers, the variance explained by this regression spline was relatively small, and the global anthropogenic Hg supply has remained constant since the mid-1990s at around 3500 tonnes annually (UNEP, 2013).

Life-history strategy may influence exposure to Hg, with female seabirds that breed bi-annually being less able to excrete metals during egg laying (Ackerman et al., 2016; Hindell et al., 1999; Monteiro and Furness, 2001a). Flesh-footed Shearwaters breeding in Western Australia and New Zealand may not breed annually (Lavers et al., 2019; Waugh et al., 2014), and therefore may not have the same opportunities to depurate Hg into eggs. Mercury concentrations did not change over time, which may be surprising given the increases observed in other studies (Bond et al., 2015; Evers et al., 2014; Thompson et al., 1993a; Thompson et al., 1992; Vo et al., 2011) which may be a function of the low number of samples early in the time series. Flesh-footed Shearwaters have experienced considerable trophic shifts since the mid-19th century, including a decrease of one trophic level, and trend towards increased dietary breadth in Western Australia (Bond and Lavers, 2014), and had the lowest contemporary concentrations of Hg (Bond and Lavers, 2011). However, feather Hg concentrations during 1946-2011 (8.241 ± 0.944 µg/g; Figure 1) are comparable to adult Flesh-footed Shearwaters sampled in Western Australia in 2008 (6.038 ± 3.998 µg/g) (Bond and Lavers, 2011). Flesh-footed Shearwaters' reduction in trophic position may have been a contributory factor in their unchanged feather Hg concentrations (Bond and Lavers, 2014). Given that the ultimate source of Hg is dietary, a detailed examination of Flesh-footed Shearwater diet and prey Hg across its breeding range would help elucidate the reasons for this pattern.

Lead

Pb was added to gasoline as an anti-knocking agent in the early 20th century, before being phased out in many countries less than 100 years later because of the negative environmental effects of Pb in automotive emissions (Seyferth, 2003; Wilson and Horrocks, 2008). Like Hg, Pb is also largely acquired through birds' diet, and binds to keratin and other proteins rich in sulfhydryl groups (Burger and Gochfeld, 2000a; Goede and de Bruin, 1984). In birds, high concentrations of Pb are associated with neurological and developmental impairment (Burger and Gochfeld, 2000a), particularly when feather concentrations exceed 4 µg/g (Burger, 1993; Burger and Gochfeld, 2000a), and local sources can dramatically affect Pb concentrations (Scheifler et al., 2006). More than a third (65/165; 37%) of shearwaters sampled had feather Pb concentrations above this level (Figure 1). The peak identified in the late 1940s and early 1950s does correspond with the rapid increase in leaded gasoline consumption (Nriagu, 1990; Seyferth, 2003), though as with Hg the r^2 of the regression spline was not high.

While Pb contamination has the potential to negatively affect the health and reproductive fitness of individual shearwaters, concentrations are decreasing over time (Table 1). The decrease we observed in shearwater feather Pb mirrors the declines in atmospheric Pb following the reduction in Pb as an additive in gasoline. Concentrations in *Sardinops sagax* muscle from the Arabian Sea, adjacent to shearwaters' over-wintering grounds (Lavers et al., 2019) were also relatively low (0.005 µg/g; Tawfik, 2013).

On offshore islands and in remote areas, even small populations of migratory species (e.g., salmon, seabirds) can transport significant quantities of hazardous contaminants via their guano (Evenset et al., 2007; Sun and Xie, 2001). As anthropogenic contamination of the marine environment increases, so, too, do inputs from the ocean to the land. Cd concentrations in Flesh-footed Shearwaters increased 1.5% per year during 1900-2011 (Table 1), suggesting guano deposition on breeding islands in Western Australia may be a previously undocumented source of chemical pollution.

Archival samples allowed us to frame contemporary ecotoxicological results in an historical context, which provided insight into changes in the pressures faced by Flesh-footed Shearwaters over the last century. Observing changes in ecosystems over such periods is challenging, as perceived baselines shift over time (Blight et al., 2015; Papworth et al., 2009). By using dated museum specimens, researchers can begin to examine historical changes in ecosystems using archived material and inform modern conservation priorities and actions.

While this study has identified temporal trends in metal concentrations, it has also highlighted a lack of information on the diet and foraging behaviour of Flesh-footed Shearwaters and population level effects from metals exposure. Between 17-72% of the shearwaters sampled for this study exceeded thresholds for Cd, Hg, or Pb. Chemical pollutant levels may be an additional stressor on the Western Australian Flesh-footed Shearwater population, which has a low annual adult survival rate (0.634-0.835; Lavers et al., 2019) or on other populations which are declining across the species range (Jamieson and Waugh, 2015; Lavers, 2015; Reid et al., 2013).

Conclusions

Flesh-footed Shearwaters have shown contrasting trends in Cd, Hg, and Pb over the 20th and early 21st centuries, driven by several factors. Concentrations of some trace elements, namely lead, may be sufficiently high to cause adverse effects, and future work should investigate this further. Our understanding of the context of contemporary contamination has been improved through examining samples from museums and biological archives.

Data availability

Data are available on figshare: https://doi.org/10.6084/m9.figshare.12076704

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Tables

Table 1. Robust regression analyses of metals in Flesh-footed Shearwater feathers. Reported values are those used in other assessments of contaminants over time in biota (AMAP, 2011).

Element	n	Number of years (range)	% increase per year (95% CI)	Years required	Lowest detectable change (%)	Power of time series (%)
Cadmium	140	36 (1900-2011)	+1.5 (+0.6, +3.0)	29	7.1	99
Mercury	137	23 (1946-2011)	-0.3 (-2.1, +1.5)	21	8.3	94
Lead	165	36 (1900-2011)	-2.1 (-3.2, -1.0)	26	76	100

Figures

Figure 1 – Temporal trends in Cd (increasing), Hg (no significant change), and Pb (decreasing) in feathers from Flesh-footed Shearwaters from Western and South Australia. Blue lines are regressions with standard errors in gray (Table 1), and the dashed red lines are concentrations of concern (see Discussion). Data are log-transformed.

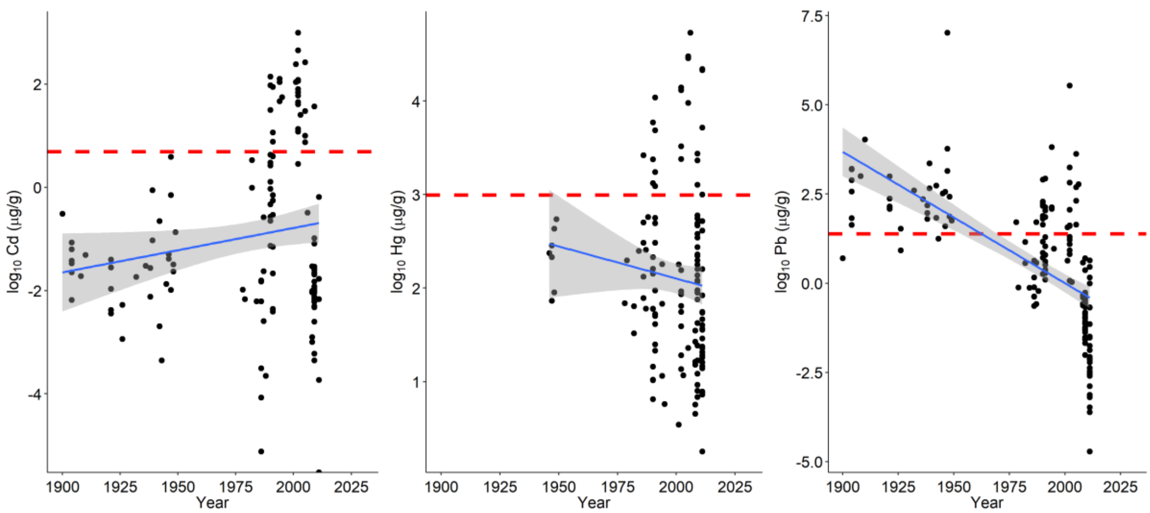

Figure 1.

Biological archives reveal contrasting patterns in trace element concentrations in pelagic seabird feathers over more than a century

Alexander L. Bond and Jennifer L. Lavers

Supplemental Material

Table S1. We achieved high recovery of two keratin-based reference materials using inductively coupled plasma mass spectrometry (ICP-MS) to measure trace element concentrations in Flesh-footed Shearwater feathers. Data are presented as the mean ± SD % recovery relative to the mean certified concentration in μg/g (ppm). Table reproduced from Lavers et al. (2013).

Reference Material (n)	**Element**	**Certified Concentration**	**Measured Concentration ± SD**	**Mean % Recovery**
6H-09 (8)	Cd	0.24	0.24 ± 0.05	100
	Hg	4.49	4.58 ± 1.81	102
	Pb	14.8	14.9 ± 0.7	100
7H-09 (8)	Cd	1.7	1.9 ± 0.1	109
	Hg	3.78	3.38 ± 1.28	89
	Pb	5.28	5.92 ± 0.75	112

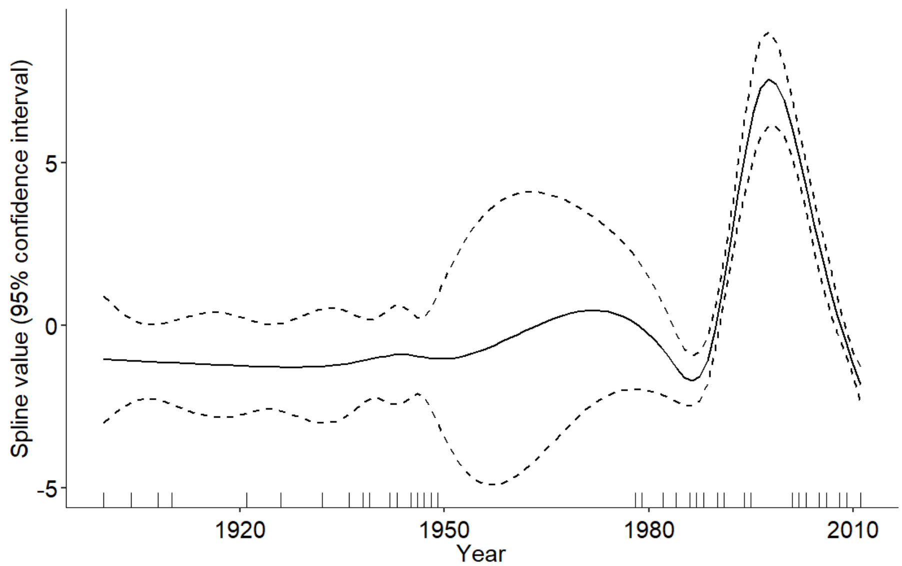

Figure S1 – The cubic regression spline of a general additive model of cadmium concentrations in Flesh-footed Shearwater feathers over time.

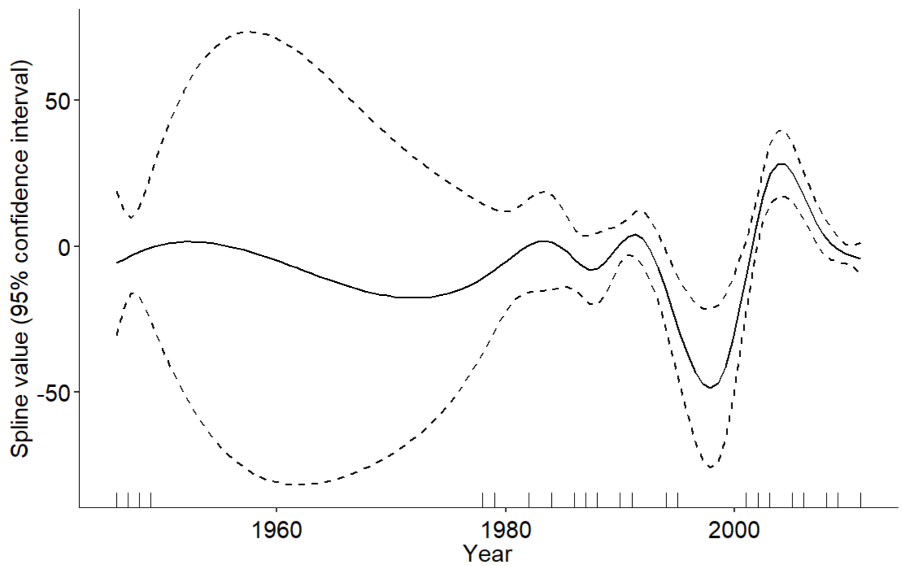

Figure S2 – The cubic regression spline of a general additive model of mercury concentrations in Flesh-footed Shearwater feathers over time.

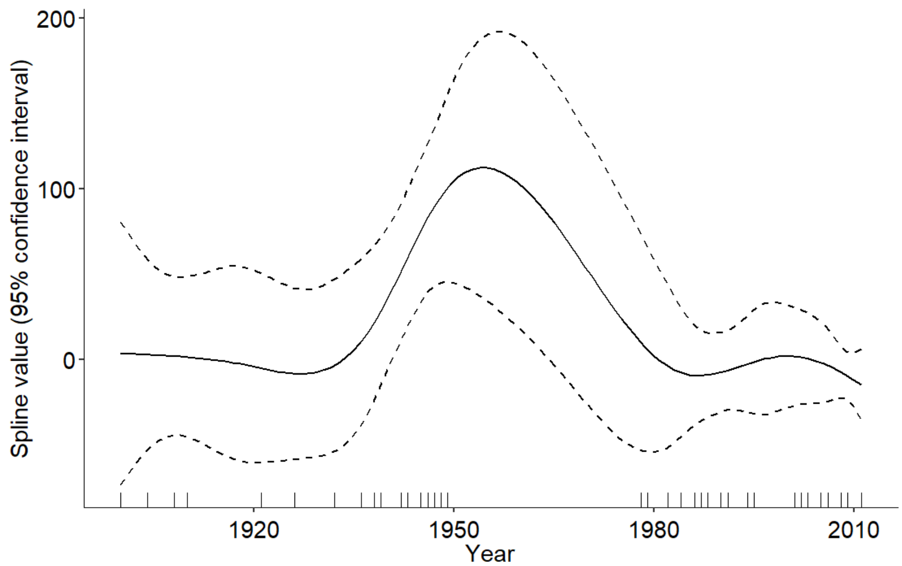

Figure S3 – The cubic regression spline of a general additive model of lead concentrations in Flesh-footed Shearwater feathers over time.